A soil ingestion pilot study of a population following a traditional lifestyle typical of rural or wilderness areas

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ABSTRACT

The relatively few soil ingestion studies underpinning the recommended soil ingestion rates for contaminated site human health risk assessments (HHRAs) that have been conducted to date assessed soil ingestion in children living in urban or suburban areas of the United States, and to a lesser extent, Europe. However, the lifestyle of populations living in North American urban and suburban environments is expected to involve limited direct contact with soil. Conversely, many populations, such as indigenous and Aboriginal peoples residing in rural and wilderness areas of North America and worldwide, participate in activities that increase the frequency of direct contact with soil. Qualitative exposure of Aboriginal populations inhabiting wilderness areas suggest that high levels of soil ingestion may occur that are many times greater than those recommended by regulatory agencies for use in HHRAs. Accordingly, a study of subjects selected from a wilderness community in Canada was conducted using mass balance tracer methods to estimate soil ingestion and the results compared with previous soil ingestion studies and regulatory guidelines for the soil ingestion rates used in HHRA of contaminated sites. A pilot study of 7 subjects living in the Nemiah Valley of British Columbia was conducted over a 3-week period. The mean soil ingestion rate estimated in this study using the 4 elemental tracers with the lowest food-to-soil ratios (i.e., Al, Ce, La, Si), was observed to be approximately 75 mg d^{-1} (standard deviation 120 mg d^{-1}), the median soil ingestion rate was 50 mg d^{-1}, and the 90th percentile was 211 mg d^{-1}. These soil ingestion rate estimates are higher than the soil ingestion estimates currently recommended for HHRAs of adults, and higher than those obtained in most previous studies of adults. However, the estimates are lower than the earlier qualitative assessments of subsistence lifestyles.

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1. Introduction

1.1. Background

The predominant exposure pathway for most heavy metals and non-volatile contaminants in human health risk assessment (HHRA) of contaminated sites is via the direct ingestion of soil. Accordingly, the soil ingestion rate values selected in HHRA are a major contributor to soil quality guidelines developed for assessing the health impacts of proposed industrial sites and/or for remediating existing sites contaminated from historical industrial activities. Soil ingestion may occur through the inadvertent ingestion of soil or dust particles that adhere to food, objects and hands, or the deliberate ingestion of soil (i.e., soil pica and geophagy), which is considered to be relatively uncommon in the general population (EPA, 1997). However, geophagy, can be a relatively common practice in indigenous peoples on all continents (Simon, 1998). Soil ingestion can also result from the inhalation of soil particles, typically between 3 and 10 μm particle size, that become trapped in the mucous linings of the nasopharyngeal tract, bronchi, and bronchioles, and, then cleared by mucociliary action and swallowed (Plumlee and Ziegler, 2006). Several studies have been conducted to estimate inadvertent soil ingestion in humans employing the following methodologies (EPA, 1997; 2009):

a) The "tracer element" method, where specific elements commonly found in soil are measured in excreta (e.g., feces and urine) and soil, and these values are used to calculate the mass of soil ingested. These studies are termed "mass balance tracer" studies when the soil ingestion calculation accounts for tracers in food and medicine.
b) The "biokinetic model comparison" method, where a biokinetic model of an element (e.g., lead) is used to calculate the mass of soil ingested given the measured concentration of the element in the blood of a subject.
c) The "survey response" method, where questions regarding the frequency of mouthing behavior and ingestion of non-food items

are used together with tracer (or other) study results to estimate soil and dust ingestion rates.

d) Qualitative/semi-quantitative assessments, where the types and frequency of specific behaviors are observed in subjects and quantitative data from other studies (e.g., soil/dust adherence to hands) are used to infer a soil ingestion rate.

A critical review of early soil ingestion studies employing survey-response and qualitative/semi-quantitative assessments and mass balance tracer methods was completed by Doyle et al. (2010). These studies yielded typically high (i.e., up to grams per day) soil ingestion estimates and the estimating methods used were concluded to be not sufficiently robust to establish defensible clean-up criteria for contaminated sites. Further, these methods have not been validated quantitatively. Biokinetic models have provided soil ingestion estimates of 4 mg d^{-1} depending based on the concentration of lead in blood samples taken from children (deSilva, 1994). However, this method depends on reliable bioavailability and pharmacokinetic data for the tracer element used, which are often scarce and variable based on the speciation of the contaminant of concern and condition of the subjects digestive system and an accurate assessment of time exposed to the contaminant of concern. Moreover, the method is highly intrusive, requiring collection of blood, which would be impractical in a remote field setting. Mass balance soil ingestion studies have been developed and validated that have progressively improved such that the normal soil ingestion rates for children and adults have been substantially reduced, and the precision of the estimates has been increased. Accordingly, a mass balance tracer approach was selected to estimate soil ingestion in this study.

Recommended rates of inadvertent soil ingestion are provided by regulators to facilitate the development of exposure assessments in HHRA of contaminated sites. Inadvertent soil ingestion does not include the intentional ingestion of soil, as in the case of soil pica or geophagy. The key primary mass balance tracer studies (i.e., studies that have generated primary soil ingestion data) that were used by the United States Environmental Protection Agency (EPA) to develop recommended values for inadvertent soil ingestion include 3 cohort studies of children living in suburban households in the United States, where tracer levels in food were measured directly (Calabrese et al., 1989, 1997; Davis et al., 1990), and 1 cohort study of children in hospitals, daycare centers and campgrounds in the Netherlands, where the tracer loading in excreta of the hospitalized children was used as a proxy measure of daily dietary intake of tracers for the other children in the study (van Wijnen et al., 1990). Other relevant studies that were considered in the development of soil ingestion guidelines include 2 pilot mass balance tracer studies of adults that ingested a known amount of soil (Calabrese et al., 1990; Stanek et al., 1997), a tracer element study of a cohort of children where tracer intake in foods was not accounted for in the soil ingestion calculations (Binder et al., 1986), and a pilot study of children (Clausing et al., 1987) using the same methods as van Wijnen et al. (1990). The recommended inadvertent soil ingestion rates for use in HHRAs made by regulatory agencies are largely derived from the aforementioned studies and range from 80 mg d^{-1} (Health Canada) to 150 mg d^{-1} (Netherlands National Institute of Public Health and the Environment or RIVM) for toddlers (6 months to 4 years of age), and from 20 mg d^{-1} (Health Canada and World Health Organization) to 60 mg d^{-1} (RVIM) for adults. The EPA recommended soil ingestion rates for use in HHRAs for toddlers and adults are 100 and 50 mg d^{-1}, respectively.

The above recommendations are based largely on a few quantitative assessments of relatively large numbers of children, augmented by smaller studies of adults, living in suburban and urban locations under controlled situations. As such, they are not necessarily representative of populations living in rural or wilderness areas with occupations or lifestyles that increase the likelihood of greater soil ingestion (Doyle et al., 2010). Moreover, there have been no quantitative soil ingestion studies of a Canadian population (Wilson Consulting, Meridian Environmental, 2006). Soil ingestion rates for receptors living in environmental conditions typical of rural or wilderness regions characteristic of many regions of Canada, or that are participating in activities that may be more vulnerable to soil ingestion, are normally assigned a soil ingestion rate from an upper quantile (e.g., upper 90 percentile) of the distribution of soil ingestion rate estimates generated from the limited number of studies of children and adults completed to date. Default soil ingestion rates for HHRA are not derived from mass balance soil ingestion studies of adults participating in activities that may be vulnerable to high soil ingestion. For example, the EPA recommends a default soil ingestion rate of 330 mg d^{-1} for a construction worker who is vulnerable to increased exposures resulting from soil-disturbing activities such as site excavation or vehicle traffic on unpaved roads (EPA, 2002). The 330 mg d^{-1} value represents the 95th percentile of the adult soil ingestion rates reported by Stanek et al. (1997). Furthermore, this soil ingestion rate of 330 mg d^{-1} has also been assumed for United States' military personnel during training or deployments (USAPHC, 2010). Moreover, Harper et al. (2002, 2005) recommended a soil ingestion rate of 400 mg d^{-1} for Aboriginal peoples following subsistence lifestyles in the Plateaus of the North-western United States. This value is an upper bounding ingestion estimate recommended by the EPA for children, with the additional assumption that traditional subsistence activities will have similar soil contact levels to that of construction and utility workers or deployed military personnel. Thus, populations participating in activities vulnerable to enhanced soil ingestion may be under protected if HHRAs of contaminated sites use the soil ingestion rates currently recommended for adults and children.

This pilot study is directed at obtaining the first quantitative soil ingestion rates, using mass balance tracer methods, of 7 subjects selected from a Canadian population following a traditional lifestyle typical of rural or wilderness communities. The purpose is to determine if soil exposure of rural or wilderness communities via the ingestion pathway is greater than the ingestion values developed for the population at large, which have been used to underpin HHRAs and regulatory decisions pertaining to contaminated sites.

1.2. Study area

The study area is located in the Chilko River watershed in the Cariboo Forest Region of British Columbia and within the traditional lands of the Xeni Gwet'in First Nation approximately 230 km west of Williams Lake (Fig. 1). The traditional lands of the Xeni Gwet'in First Nation generally represent the land enclosed by Chilko Lake (Tsilhqox Biny) and the Chilko River (Tsilhqox) to the west, and the Taseko River (Dasiqox) to the east, with a southern boundary running through the Nemiah Valley (Xeni) (Supreme Court of British Columbia, 2007). The area encompasses plateau, glaciated mountains and transition zones of the Chilcotin and Pacific Ranges.

The soil ingestion study was conducted at the following locations:

- Nemiah Valley (Fig. 1 — Location "A")
- Henry's Crossing (Fig. 1 — Location "B")
- Farwell Canyon (Fig. 1 — Location "C").

The surficial geology and geochemical characteristics of the study area are a complex mixture of bedrock, of both marine sedimentary and volcanic origins, overlain within the Nemiah Valley by Pleistocene gravel, glacial tills, silt and clay (GSC, 1935; Schiarizza et al., 1995; Umhoefer et al., 2002). The sedimentary rock (i.e., sandstones) in the region is typically quartz poor, ranging from lithanarites to feldspathic litharenites to lithic arkoses. Soils originating from plagioclase andesite and arkose sandstones are predominated by feldspar lithic components and a chlorite cement $((Mg,Fe,Al)_6 (Si,Al)_4O_{10}(OH)_8)$. Plagioclase minerals are composed of sodium aluminum silicates
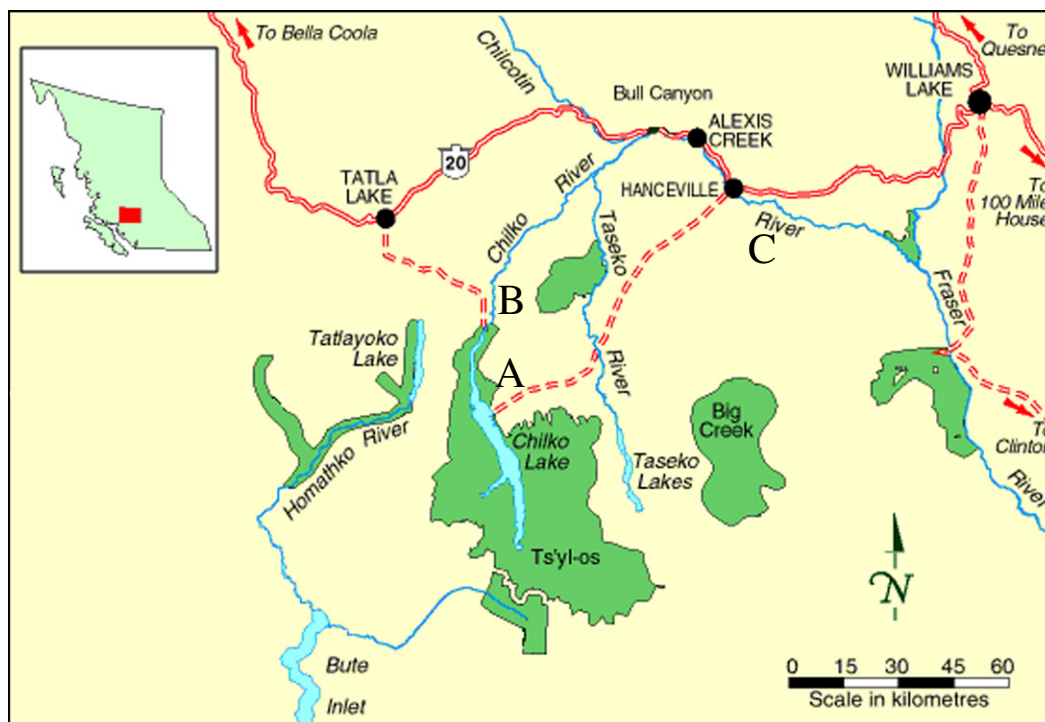
Fig. 1. Map of the study area with an inset showing the location of the study area within British Columbia. Shaded areas denote BC Provincial Parks and protected areas. Solid lines denote paved road and hatched lines denote dirt or gravel road.

($NaAlSi_3O_8$), such as albite, or by calcium aluminum silicates ($CaAl_2Si_2O_8$), such as anorthite (Leet and Judson, 1971). The study area has a moderate continental climate with cold winters, warm summers and relatively low levels of precipitation. As such, the conditions in the study area are conducive to the production of dust clouds resulting from vehicular traffic, winds and/or activities that stir up soils (e.g., horse and cattle movements).

The study was conducted in cooperation with the Xeni Gwet'in First Nation community residing in the traditional lands in the Nemiah Valley. Formerly known as the Nemiah Band, the Xeni Gwet'in is 1 of 6 Tsilhqot'in First Nation communities residing in the Chilcotin Plateau and Chilcotin Mountain Range in British Columbia. An engineered gravel and dirt road was constructed by the Canadian Army Corps of Engineers in 1973 from Konni Lake to highway 20 at Hanceville. To sustain themselves before the road was constructed, the Xeni Gwet'in ran cattle and trapped wildlife through the winter, and gardened, hunted and fished in the spring and summer months (Xeni Gwet'in, 2011). The Nemiah Valley has ecosystem characteristics and habitats that support the traditional lifestyle of the inhabitants and the Xeni Gwet'in First Nation government has clearly articulated the importance of a traditional lifestyle to the well-being of their community.

2. Methods

2.1. Mass balance soil ingestion study design

The study involved 7 adult (i.e., greater than 20 years old) volunteer subjects, 4 days per week over a 3-week period between August 16 and September 2, 2011. The study was conducted at this time of year because it is representative of dry and windy conditions conducive to enhanced soil ingestion and was also during the annual salmon migration period when traditional fishing activities are conducted. Four subjects were community members of the Xeni Gwet'in and 3 were not. A formal agreement with the subject community and

approvals from the Health Canada and the University of Ottawa research ethics review boards were required before the research could begin. A Memorandum of Understanding was signed with the Xeni Gwet'in First Nation Government that secured the participation of the subject community, and approvals to conduct the research were also obtained from the University of Ottawa and Health Canada Research Ethics Review Boards (REBs). All participants in the study were briefed on the objectives, the scope of their participation and their role in the study, and signed a consent form in accordance with research ethics protocols. The subjects were also briefed each week on the protocols for providing fecal samples.

During Week 1 of the study, 2 of the 7 subjects participated in the traditional fishery, 1 subject at Location A and the other subject at Location C (Fig. 1). During Weeks 2 and 3 of the study period, 6 of the 7 subjects participated in establishing a commercial First Nations fishery on the Chilko River at Location B (Fig. 1). Five of the 7 subjects participated in both Weeks 2 and 3, 1 subject participated in Week 2 and the remaining subject in Week 3. Daily activities included clearing deadfall from spawning streams, collecting Sockeye salmon using traditional methods, such as "dip nets" or seine nets along the shore, weighing, bleeding and cleaning each fish, and storing the catch in a mixture of brine and ice. The late afternoon and evening involved scouting of new dip net locations by hiking up the shore of the river or fishing for Sockeye and Chinook salmon with rod and reel, in addition to routine camp activities (e.g., eating and clean-up, collecting and cutting firewood, etc.). No other demands were placed upon the subjects participating in the study, and several participants took the opportunity to fish with rod and reel and/or hunt in the evenings. The activities included in the study (i.e., traditional fishing, attending gatherings, etc.) were selected because, based on initial discussions with community leaders and the information collected in the ethno-cultural survey, they were typical of traditional community activities.

Soil ingestion was estimated using a mass balance tracer methodology. Reliable mass balance tracers are not readily absorbed in the gastrointestinal tract, are ubiquitous in soils and are generally found

in low concentrations in food. The tracers selected for this study were Al, Ba, Ce, La, Mn, Si, Th, Ti, U, V, Y and Zr because they generally meet these criteria and have been used in previous studies (Calabrese et al., 1989; 1997). Th and U were also included as tracers to support a separate study evaluating the feasibility of using naturally occurring radionuclides as mass balance tracers for estimating soil ingestion. The daily soil ingestion for each subject was calculated from Eq. (1).

$$S_a = \frac{F_c \times F_a}{S_c} - \frac{I_c \times I_a}{S_c} \qquad (1)$$

where:

S_a is the soil ingested (g)
F_c is the concentration of tracer element in feces ($\mu g\, g^{-1}$)
F_a is the mass of feces (g)
I_c is the food concentration for tracer element ($\mu g\, g^{-1}$)
I_a is the mass of food ingested (g)
S_c is the concentration tracer in soil ($\mu g\, g^{-1}$).

A daily soil ingestion rate was calculated for each subject using the food intake on Day 0, Day 1 and Day 2 (and Day 3 for subjects in Week 1). A 24 h transit time was assumed, and the fecal output (i.e., the F_a, and F_c parameter values from the analyses of the daily fecal samples) was obtained for Day 1, Day 2 and Day 3 (and Day 4 for subjects in Week 1) of each study week. It is noted that gastrointestinal transit time will vary substantially between individuals in response to differences in gender, age and diet. The 24 h transit time assumption is close to a median aggregate of gastric, small intestinal and colonic transit times of 28.5 h measured by Madsen and Jensen (1989) and allows for a consistent collection of fecal samples from each subject on a daily basis. Logistical constraints presented by conducting the study in a remote wilderness area with adults precluded the collection samples of urine or sweat. Excluding tracer levels in these excreta from the mass balance calculations will negatively bias the soil ingestion estimates. However, this bias is expected to be small given the low bioavailability of the tracers employed in this study. Food intake (I_a) was calculated as the product of the number of portions of each food type ingested (recorded in daily food ingestion logs) and the pre-weighed portion size. The portion weights were converted to dry weight by dividing the wet weight by a dry weight concentration factor derived in the laboratory for each food item analyzed. The types and quantity of medications taken by each subject were also recorded. I_c was derived from the analysis of the food item types. S_c was obtained from the mean tracer level in <63 μm particle size soil obtained from the location where the subjects were working during that particular study week. The <63 μm particle size was used because this fraction best represents the fraction that adheres to hands, and is thus most likely to be ingested (Doyle et al., 2010).

2.2. Soil sample collection and sample processing

Soil samples were collected at the 3 locations identified in Fig. 1. Samples were collected by scraping the surface soil from a 10 cm by 10 cm area to a depth of 2 cm, yielding approximately 200 mL of sample. At Location B and Location C, 5 samples were collected in a cross formation 25 m apart. A soil sample was collected at each of the 3 areas in the Nemiah Valley (Location A) where work was performed. Seven soil samples were also collected, as per the aforementioned methods, along the main travel route transecting the study area. The samples were collected along the main road into the Nemiah Valley beginning at the Vedan River Crossing in the most Eastern portion of the study area, through to Konnie Lake to the eastern shore of Chilko Lake, then north along the east side of the lake to Henry's Crossing, then crossing the Chilko River to Choelquot Lake located

in the most western portion of the study area. All soil samples were collected in WhirlPak™ plastic sample bags, labeled and shipped to the laboratory in Ottawa.

In the laboratory, soil samples were oven-dried for approximately 24 h at 90 °C, lightly de-consolidated in a ceramic mortar with a rubber pestle and sieved into >2 mm, <2 mm–>250 μm, 250 μm–>63 μm, and <63 μm particle size fractions. A 1 g subsample of the <63 μm soil fraction was transferred to a 20 mL glass vial and stored until analysis.

2.3. Food and water sample collection and sample processing

In previous mass balance studies, duplicate samples of entire meals were collected from each subject home. Given that this study was conducted in a remote location with only limited secure storage space (i.e., to protect the samples from animals), storage of duplicate meals and snacks for all subjects over the study period was considered impractical. Instead, tracer ingestion was calculated by quantifying the foods consumed for each subject for each day and analyzing matching samples of each food item for tracer concentration. It is anticipated that this difference in methodology will result in only a minor increase soil ingestion estimate uncertainty. In an unpublished study, 28 subjects at a buffet lunch were asked to provide duplicate plates of food for analysis, simulating how previous studies determined tracer intake with food. The mean difference in meal weight and duplicate meal samples was observed to be 9% of total weight compared to an overall coefficient of variation 12% measured for portion sizes in this study. Moreover, food consumption by was closely monitored for the entire duration of this study and it is anticipated that bias resulting from unreported food consumption by subjects will be less than for studies requiring unsupervised subjects to comply with study protocols.

All foods (breakfast, lunch, dinner and snacks) were provided to the subjects beginning on lunch Day 0 and ending with lunch Day 4 of each week (i.e., the time spent in the field camp). The types of meals planned, food items provided and preparation methods were kept the same for each week. Duplicate samples of food types provided to the subjects were collected when provisions were purchased and, frozen where necessary, then shipped to the laboratory. The water consumed at Location A was obtained from Chilko Lake, and the water consumed at Location B was obtained from the Chilko River at Henry's Crossing. The water consumed at Location C was obtained from Location A, and transported to the field camp near Farwell Canyon. Approximately 2.5 L samples of water were collected from both locations in 1 L polyethylene bottles, shipped to the laboratory then acidified to a pH<2.0 with concentrated HCl.

Average weights of the specific food portions (e.g., slices of meat, servings of potato, cups of tea) served were predetermined in the laboratory and/or in the field, and the number of servings of each food item was carefully logged for each subject for each meal, including snacks. Food consumption for each subject was tracked from lunch Day 0 until the end of Day 3 (i.e., including dinner and evening snacks). Each subject was also interviewed to determine the types of foods eaten for breakfast on Day 0 relative to the servings provided by the study.

Samples of foods provided to the study subjects were procured and retained for analysis. Food samples were ashed at 500 °C for 9 h and weighed. A 1 g sub-sample was collected of all food types and water samples and transferred to 20 mL glass vials and stored until analyzed.

2.4. Fecal sample preparation

Daily fecal samples were obtained from each subject from Day 2 until the end of Day 3. Portable commodes and pre-labeled and pre-weighed sample containers (Fisher Scientific autoclavable

polypropylene biohazard sample bags, catalog number 01-826-5) were provided to the subjects for the collection of fecal samples. Samples were sealed by the subject with 15 cm cable ties and placed on ice in a large cooler lined with a polypropylene bag. Each day, the subjects reported the time(s) the fecal sample(s) that was/were provided, and the sample bag number(s). For the 4-day sampling period, the fecal sample bags were frozen and transferred into a second bag. At the end of the study, the frozen samples were sealed in large sample coolers and stored in a freezer until they were shipped by air cargo under refrigeration. Upon receipt, the fecal samples were transferred into dedicated freezers for storage until further processing.

All fecal sample handlings in the laboratory were conducted under a fume hood, except when samples were being transferred from freezers to the fume hood or a muffle furnace. Samples were dried in the sample bags in an oven (enclosed in the fume hood) for approximately 48 h at 90 °C then re-weighed. Fecal matter was then removed from the sample bags into pre-weighed crucibles, ashed at 500 °C for 9 h and weighed. The ashed fecal samples were then consolidated, as required, into composite samples representing each study day for each subject. A 1 g sub-sample was removed from each 8 mL tube and transferred to 20 mL glass vials and stored until analyzed.

2.5. Analytical methods for tracers

Analysis of the tracer elements (Al, Ba, Ce, La, Mn, Si, Th, Ti, U, V, Y and Zr) was performed by a commercial laboratory accredited by the Canadian Association for Laboratory Accreditation Inc. to ISO/IEC 17025:2005. For the analysis of Al, Ba, Ce, La, Mn, Th, Ti, U, V, Y and Zr, samples were digested using EPA Method 3052 (i.e., digested in concentrated nitric acid and hydrofluoric acid using microwave heating). Digested samples were then analyzed by inductively coupled plasma mass spectrometry (ICP/MS) for the metal tracers. Total Si was determined by sodium peroxide fusion followed by inductively coupled plasma optical emission spectrometry (ICP/OES).

2.6. Statistical analysis

Statistical analyses were conducted using JMP® or Microsoft Excel™ software. Analysis of variance (ANOVA) was used to compare normal or near normal distributions with similar variance and their means compared using Tukey–Kramer Honestly Significant difference (HSD) method and with the Kruskall–Wallis test when the distributions were not normally distributed. Normality of the distributions was determined using the Shapiro–Wilk test. When distributions had different variances, the Welsh ANOVA was used to determine if differences in the distributions were statistically significant. Differences in variances were determined by the Levene test.

3. Results

3.1. Study conditions

The conditions during the 3-week study period between August 16 and September 2, 2010 were generally warm during the day, cool at night, and dry. The weeks preceding the study were characterized by a series of major forest fires in the area that continued to burn during Week 1, and subsequently subsided in Weeks 2 and 3. However, the active fires were at approximately 100 km or more of mountainous terrain from the actual study sites. The mean daily temperature, mean daily maximum temperature and monthly precipitation measured at the Environment Canada Tatlayoko Lake weather station (approximately 21 km west of Henry's Crossing) for the month of August were 14.6 °C, 24.5 °C and 13.4 mm, respectively (Environment Canada, 1971–2000). Winds were moderate and were

reported to be less than 30 km h^{-1} at Tatlayoko Lake throughout the study period.

3.2. Soil samples

The mean and standard deviation tracer levels measured in the soil sampled at the 3 soil ingestion study locations and 7 samples transecting the study area are summarized in Table 1.

3.3. Food samples and daily tracer consumption rates

It has been shown that the uncertainty of soil ingestion estimates using the mass balance tracer method is greatest for tracer elements with higher food/soil (F/S) ratios, defined as the mass of the tracer element ingested from food over a one day period divided by the mass of the tracer element in 1 g of soil (Calabrese and Stanek, 1993). Accordingly, to identify the most reliable tracers, daily F/S were calculated for each subject based on the calculated daily tracer consumption by each subject, and the mean tracer concentrations measured in soils. The mean, standard deviation and coefficient of variation (CV) of tracer intake by all subjects expressed as mass of tracers consumed and as the F/S ratio are provided in Table 2. It is noted that the number of food sample types that were below the minimum detection limit (MDL) for V was high, with 17 out of a total of 19 analyses below detection. For samples where the tracer levels were less than the MDL, a value of 1/2 of the MDL was used in the calculation of daily tracer intake.

The variability of the daily intake of tracers was observed to be moderate, as reflected by CV values ranging between 27% and 52% for 8 of the 11 tracers. The highest variability was observed in the ingestion of Th and Ti tracers, with CV values of 82% and 98%, respectively. The variability in daily consumption rates for Ti were likely the result of the tracer concentrations in granola bars and doughnuts being 2 orders of magnitude higher than the Ti levels measured in the other food types. It was noted that both of these food items contained processed toppings (i.e., colored icing), and these high levels are possibly due to TiO$_2$, a common coloring agent in processed foods and consumer products. Elevated levels of Al were also observed in pancake mix and doughnuts, probably due to aluminum sulfate in baking powder, which is used as a leavening agent.

Al, V, Si, La and Ce, were observed to be the most reliable tracers, with mean F/S values of 0.06, 0.07, 0.08, 0.13 and 0.13, respectively, and CV values ranging between 47% and 67%. However, it is noted that the F/S values for V are based on only 2 analyses of food samples above the detection limit, and its value as a mass balance tracer is questionable. Thus, the 4 most reliable tracers identified in this study are Al, Si, La and Ce. Al and Si are considered 2 of the most reliable tracers that have been used in previous soil ingestion studies (Stanek et al., 2001). This observation is supported by the current study where these elements were observed to have lower F/S values than La and Ce for each study day, with the exception of 2 days, when Si had slightly higher or comparable F/S values.

3.4. Fecal samples

Fecal samples were successfully collected each day from all subjects; however, one fecal sample was lost (Subject D, Week 2, Day 3). The dry weight of daily fecal output for each subject was measured and the data are summarized in Table 3. It was observed that the total dry weight of fecal output over the study period for all subjects was approximately 3.7 kg, or approximately 26% of the total dry weight of food consumed. The mean and median fecal tracer concentrations for all subjects over the duration of the study are summarized in Table 4. The mean fecal dry weights for all subjects on Day 1, Day 2 and Day 3 of the study were observed to be 36.6 g, 45.4 g and 44.6 g, respectively. Although it was observed that the mean fecal

Table 1
Tracer levels measured in soil samples from the soil ingestion study area and across the Nemiah Valley (A is Nemiah Valley in Fig. 1, Location A; B is Henry's Crossing in Fig. 2, Location B; C is Farwell Canyon in Fig. 2, Location C; NS are samples taken every 15 km bisecting the Brittany Triangle roughly from east to west as shown in Fig. 2). SD is the standard deviation and CV is the coefficient of variation (i.e., standard deviation divided by the mean).

Sample number	$\mu g \, g^{-1}$											
	Al	Ba	Ce	La	Mn	Th	Ti	U	V	Y	Zr	Si
A-Mean	63,333	447	22.7	10.3	767	1.9	3567	0.9	123	12.0	47.7	270,000
NV-SD	27,301	5.8	1.2	0.6	115	0.1	635	0.0	23	0.0	9.8	18,200
CV	43%	1%	5%	6%	15%	6%	18%	2%	19%	<1%	21%	7%
n	3	3	3	3	3	3	3	3	3	3	3	3
B-Mean	66,800	500	23.8	10.8	980	2.1	3720	1.1	103	13.2	37.2	240,800
HC-SD	2864	25.5	0.8	0.4	76	0.1	130	0.2	7	0.8	3.1	7250
CV	4%	5%	4%	4%	8%	4%	4%	22%	7%	6%	8%	3%
n	5	5	5	5	5	5	5	5	5	5	5	5
FW-Mean	72,400	550	28.8	13.2	1022	3.1	5680	0.8	122	14.2	62.4	229,800
C-SD	5030	35.4	2.2	1.1	80	0.2	814	0.1	18	1.6	7.0	6600
CV	7%	6%	8%	8%	8%	6%	14%	11%	15%	12%	11%	3%
n	5	5	5	5	5	5	5	5	5	5	5	5
NS-Mean	72,900	430	24.0	11.3	779	2.1	3440	1.0	105	13.7	37.9	245,700
NS-SD	5400	30.6	3.4	1.8	93	0.3	710	0.2	18	2.3	11.8	5600
CV	7%	7%	14%	16%	12%	16%	21%	23%	17%	17%	31%	2%
n	7	7	7	7	7	7	7	7	7	7	7	7

dry weight was approximately 10 g lower on Day 1 compared to Days 2 and 3, the difference was not significant (ANOVA, F = 1.48, p = 0.24; Welch ANOVA, F = 1.14, p = 0.34). The increase in the daily fecal dry weight on Days 2 and 3 may be a result of increased total food intake or an increase of cellulose fiber in the diet provided to the subjects during the study.

3.5. Mass balance soil ingestion estimates

A summary of soil ingestion for each week for all subjects is provided in Table 5. The soil ingestion rate, calculated for the 4 most reliable tracers (i.e., Al, Si, La and Ce) provided in Table 6. These tracers were selected because they best met the aforementioned criteria for reliable tracer elements. Specifically, the tracers are poorly absorbed in the gastrointestinal tract (assumption "c"). absorption factors (i.e., f_1 or the mass of tracer absorbed divided by the mass of tracer ingested) used in pharmacokinetic modeling of radionuclides in humans, are 0.01, 0.005, 0.005 and 0.01, for Al, Ce, La and Si, respectively (ICRP, 1996). Furthermore, Al and Si, originating from Plagioclase minerals, (i.e., $NaAlSi_3O_8$ and $CaAl_2Si_2O_8$) in the bedrock and soils commonly found in the study area are insoluble or only sparingly soluble in water (CRC, 1993). Moreover, these tracers were observed to have the lowest F/S ratios (excluding V) of the tracers used to calculate the soil ingestion rate.

There were difficulties with the Si analysis of 13 fecal samples due to insufficient sample. As such, there were no completed estimates for Si in these cases, resulting in no Si-based soil ingestion estimates for Subject A in the study. Given that Subject A was observed to have the highest soil ingestion rate of all subjects, based on soil ingestion

estimates using the Al tracer, the missing soil ingestion analyses may represent a negative bias in soil ingestion estimate based on the combined Al and Si tracers, and the 4 most reliable tracers. The soil ingestion rate distributions estimated for each tracer are shown in Fig. 2, and their means were observed to be significantly different (ANOVA, Tukey–Kramer HSD, p = 0.003), with soil ingestion estimates obtained using La being significantly higher than Al and Si (p < 0.003), but not Ce.

4. Discussion

This study has largely been directed at addressing the lack of quantitative soil ingestion data to support HHRA exposure scenarios for populations living in rural or wilderness areas of Canada. To this end, the study has assessed soil ingestion that would be incurred participating in wilderness camping and activities related to traditional fishery practices of a First Nation in the interior of British Columbia. It is important to note that the soil ingestion estimates in this study are based on the inadvertent ingestion of soil from participating in what were deemed to be "intermediate contact" activities, such as camping, hunting or fishing, and did not include ingestion related to "high contact" activities and/or ingestion of soil in traditional foods. Thus, soil ingestion rates for First Nations inhabitants of wilderness areas who engage in traditional subsistence activities could be higher. Moreover, the soil ingestion estimates in this study do not include the contribution from ingesting soil adhering to locally-sourced and/or -preserved food, and this contribution would be expected to increase soil ingestion rate in community members that consume traditional foods. Although these activities are not necessarily representative of those that would result in the highest rates of soil ingestion (i.e.,

Table 2
Mean, standard deviation (SD) and coefficient of variation (CV) for daily tracer consumption rates and food/soil (F/S) ratios during the soil ingestion study for all soil ingestion study subjects.

Daily tracer ingestion	Tracer consumption ($\mu g \, d^{-1}$)										
	Al	Ba	Ce	La	Mn	Si	Th	Ti	V	U	Zr
Mean	4860	519	3.6	1.7	2360	22,300	2.2	1780	8.4	1.3	17.2
SD	1970	190	1.2	0.4	1170	10,800	1.8	1730	4.4	5.4	6.2
CV	41%	37%	34%	27%	49%	48%	82%	98%	52%	41%	36%
F/S ratio											
Mean	0.06	0.91	0.13	0.13	2.18	0.08	0.79	3.79	0.07	1.21	0.34
SD	0.03	0.49	0.07	0.06	1.41	0.05	0.75	4.19	0.05	0.70	0.18
CV	54%	54%	52%	47%	65%	64%	95%	110%	67%	58%	53%

Table 3
Mean, standard deviation (SD), coefficient of variation (CV), median, and upper and lower 95% confidence limits of the mean of the daily fecal output dry weight for each soil ingestion study subject.

Subject	n	Dry weight (g)					
		Mean	Std dev	CV	Median	Lower 95%	Upper 95%
A	6	43.2	9.0	21%	44.4	33.8	52.7
B	6	32.7	19.1	58%	30.0	12.6	52.8
C	6	40.7	13.0	32%	39.3	27.1	54.3
D	5	32.0	11.2	35%	31.9	18.1	46.0
E	3	41.4	6.3	15%	42.7	25.6	57.2
F	7	39.9	21.2	43%	51.4	20.4	59.5
G	10	49.8	15.7	31%	51.0	38.6	61.0

Table 4
Mean, standard deviation (SD), median and number (n) of the tracer concentration in ashed daily fecal output from all soil ingestion study subjects.

	Tracer concentration ($\mu g\ g^{-1}$)										
	Al	Ba	Ce	La	Mn	Si	Th	Ti	V	U	Zr
Mean	1673	179	1.29	0.74	1271	6821	0.30	3237	4.5	0.4	4.4
SD	650	35	0.97	0.52	527	4045	0.36	3602	2.6	0.2	2.0
Median	1600	180	1.00	0.56	1200	6350	0.22	2300	4.0	0.4	4.0
n	43	43	43	43	43	30	43	43	43	43	43

Table 5
Mean, standard deviation (SD), and median of the daily soil ingestion rates calculated for each study week for all subjects.

Subject	Soil ingestion rate ($mg\ d^{-1}$)										
	Al	Ba	Ce	La	Mn	Si	Th	Ti	V	U	Zr
Week 1											
Mean	32	556	−6	46	5610	66	−768	141	76	412	−85
SD	57	1005	128	134	6619	57	1176	1020	95	963	250
Median	45	374	−5	22	2580	61	−495	515	96	542	21
Week 2											
Mean	30	594	96	152	2857	62	−530	−302	171	586	223
SD	45	588	131	127	2417	79	669	6168	144	1038	499
Median	27	434	60	117	2238	47	−400	−586	127	480	98
Week 3											
Mean	46	586	84	153	3288	22	118	−1902	79	405	69
SD	58	804	120	191	3109	77	1558	4608	157	1277	326
Median	33	589	53	158	2971	4	0.3	−1222	65	97	36

grams per day), the activities, environmental conditions, and time spent outdoors engaged in traditional activities are substantially different from the activities ordinarily encountered in urban or suburban lifestyles. Thus, it was hypothesized that soil exposure rates for the study subjects would be measurably greater than for studies of urban/suburban populations. To test the hypothesis, the results of this pilot study of 7 subjects were compared to the results of previous studies of adults and children, as well as regulatory soil ingestion rate guidelines for use in HHRA.

Table 7 compares the distribution of soil ingestion estimates reported in the key and relevant studies underpinning the soil ingestion guidelines recommended for HHRA with the soil ingestion estimates derived from the mean values of the 4 most reliable tracers in this study, and those calculated using Al and Si values. The soil ingestion data for mothers and fathers of the children that participated in the Davis et al. (1990) are also provided in Davis and Mirick (2006). Raw data from the soil ingestion study of children at the Anaconda superfund site by Calabrese et al. (1997) and the soil ingestion study of children in Washington by Davis et al. (1990) have also been included in the discussion of results from this soil ingestion study. Data from the aforementioned studies were obtained courtesy of Professor Ed Stanek, University of Massachusetts, Amherst (UMass, 2005).

The mean soil ingestion rate of approximately 75 mg d⁻¹, determined using the 4 most reliable tracers, was much higher than the 6 mg d⁻¹ rate calculated with the Best Tracer Method (BTM) employed in the Stanek et al. (1997) study of adults. Further, the soil ingestion rate of approximately 50 mg d⁻¹, calculated using the Si tracer in this study, was greater than the 23 mg d⁻¹ and 26 mg d⁻¹ calculated using Si for Mothers and Fathers, respectively, in the Davis and Mirick (2006) study of families. Conversely, the 37 mg d⁻¹ calculated using the Al tracer in this study was less than the 92 mg d⁻¹ and 68 mg d⁻¹ calculated using Al for Mothers and Fathers, respectively, in the Davis and Mirick (2006) study.

The median soil ingestion rate values using the 4 most reliable tracers, Al or Si were generally observed to be higher than median values reported in previous studies of adults, except the one instance, where soil ingestion was calculated with the Al tracer in the Calabrese et al. (1990) study. When compared to soil ingestion rates recommended by regulatory agencies for use in HHRAs, the rates measured in this study are typically higher. Moreover, the 90th percentile for the soil ingestion rates are much higher than the regulatory guidelines for adults, with values of 211, 110 and 145 mg d⁻¹ for estimates based on the 4 most reliable tracers and the Al and Si tracers, respectively. However, these regulatory guidelines are based on reasonable central estimates of soil ingestion and the 90th percentile values from this study are lower than the aforementioned rates assigned for construction workers, military personnel or populations following subsistence lifestyles.

The mean soil ingestion rate estimates calculated in this study were generally observed to be higher or comparable to estimates reported in studies of children. The mean soil ingestion rate of approximately 75 mg d⁻¹ estimated with the 4 most reliable tracers was higher than the mean rate estimated in the Calabrese et al.

(1997) of children that used Al and Si tracers, and comparable to the van Wijnen et al. (1990) study of children in a daycare center and the Davis et al. (1990) study. The mean soil ingestion values estimated in this study were observed to be substantially lower than those estimated for children in campgrounds using the LTM. However, the median values in this study were observed to be comparable to, or higher than the median values estimated in the aforementioned studies of children. Moreover, the upper 90% quantile of the distribution is higher or of the same scale as the aforementioned recommended soil ingestion central tendency rate for toddlers (i.e., 80 mg d⁻¹). Given that toddlers are considered to be more vulnerable to soil ingestion than adults in HHRAs, it would be of interest to determine if the rate of soil ingestion for children living in rural or wilderness areas and following a traditional lifestyle is within regulatory guidelines. To this end, future studies of toddlers in these populations are warranted.

The soil ingestion estimates calculated using the Al tracer for Week 1 and Week 3 of the study were normally distributed (Shapiro–Wilk W = 0.88, p = 0.17; W = 0.93, p = 0.26, respectively); however, Week 2 was not normally distributed (Shapiro–Wilk W = 0.81, p = 0.002). There were no statistically significant differences (ANOVA F = 0.43, p = 0.65; Kruskall–Wallis test χ^2 = 0.45, p = 0.80), between soil ingestion rates measured in Weeks 2 and 3 of the study, when activities related to the traditional fishery were conducted, and the estimates from Week 1 (Fig. 3). The median soil ingestion rate measured in Week 1 was observed to be 34 mg d⁻¹, compared to median values of 44 and 47 mg d⁻¹ in Weeks 2 and 3, respectively. Overall, however, the soil ingestion rates measured in this study are only incrementally greater than those observed in previous studies, and do not support the proposed notion of gram per day soil exposure scenarios for subsistence lifestyles. However, as previously noted, the soil ingestion rate estimates reported in this study are considered conservative (i.e., less than actual values) because the ingestion of soil adhering to locally sourced and traditionally prepared food and the contribution to the soil ingestion rate estimates from 'high-contact' activities were not included. Thus, more work is required to firmly establish recommended soil ingestion rates to adequately protect people practicing traditional lifestyles typical of rural or wilderness areas. To this end, soil ingestion studies for potentially higher soil contact activities (e.g., root digging, attending and/or participating in rodeos, plowing, etc.) are warranted.

The validity of soil ingestion estimates determined using mass balance tracer methods is based on the following generic assumptions (Stanek and Calabrese, 1991):

a) The tracer element is not present, or present at low concentrations, in the food, water or medicines consumed during the study.

Table 6

Mean, standard deviation (SD), coefficient of variation (CV), the upper 95% confidence limits of the mean (Upper 95%), the median, the 75th and 90th percentiles and the maximum for the distribution of daily soil ingestion estimates calculated for the 4 most reliable tracers (Al, Si, La and Ce), for Al and Si combined, and all 4 tracers combined.

Level	n	mg d^{-1}							
		Mean	SD	CV	Upper 95%	Median	75% Quantile	90% Quantile	Maximum
Al	43	36.9	51.9	141%	52.8	31	61	110	177
Ce	43	72.2	179.5	179%	112.1	51	142	217	516
La	43	132.6	158.6	120%	181.4	104	211	343	683
Si	30	49.4	73.7	149%	76.9	40	102	145	231
Al and Si	73	42.0	61.6	147%	56.4	32	89	124	231
All 4 tracers	159	74.7	119.5	160%	93.4	50	130	211	683

b) If the tracer is present in food, then there is a one-to-one correspondence between the intake of tracer from food, water and medicines, and tracer output in feces after a defined lag or transit time, thereby allowing the calculation of soil ingested by subtracting the amount of tracer contained in food (a lack of a one-to-one correspondence is termed transit time misalignment).

c) The tracer is not absorbed in the gastrointestinal tract.

d) All tracers ingested in food and medicine are accounted for (i.e., source error resulting from inadvertent and unmeasured ingestion of tracers in consumer products) is eliminated.

e) The tracer is uniformly present at high (measurable) concentrations in soils where the study is being conducted.

The lack of correspondence between tracer intake and output can be offset by selecting tracers with low F/S ratios, increasing the duration of the study or reducing the day-to-day variability in tracer intake. In this study, the uncertainty related to assumptions "a" and "b" was largely addressed by basing the soil ingestion estimates on those with the lowest F/S ratios. Given that the study duration could not be increased due to constraints in the availability of the subjects, uncertainty related to transit time misalignment was further reduced through the provision of daily food rations to study subjects that resulted in a consistent daily intake of tracers by all subjects. For example, fresh meats and vegetables were observed to have uniformly low tracer levels, and the diets provided were predominated by fresh meats and vegetables and contained a minimum of processed foods that could contain high levels of tracers. Exceptions to this were the high tracer levels measured in granola bars, doughnuts and pre-made pancake mix that were attributed to tracers in food additives (i.e., colored icing and leavening agents). Elimination of these processed foods would further decrease the variability of tracer

intake in future soil ingestion studies. Moreover, the Nemiah study subjects were not ingesting tracers listed as active ingredients or excipients in the prescription drugs they were taking during the study. The uniformly low tracer levels in foods were reflected by the relatively low CV of food intake in subjects observed over the duration of the study. For example, the CV values for the 4 most reliable tracers (Al, Ce, La and Si) used in this study were 41%, 34%, 27% and 49%,

Table 7

Selected soil ingestion rate estimates for mass balance tracer studies reported as the "key studies" underpinning the EPA soil ingestion recommendations for HHRA.

From EPA, 2009; Van Holderbeke et al., 2007; Wilson Consulting, Meridian Environmental, 2006; UMass, 2005.

Study and tracer/ method	n	Soil ingestion rate (mg d^{-1})			
		Mean	Standard deviation	Median	90th percentile
Studies of children					
Calabrese et al. (1989)					
Al	64	153	852	29	
Si	64	154	693	40	
van Wijnen et al. (1990)					
Daycare centers (LTM)[a]	162	69	286		
Campgrounds (LTM)	78	120			
Davis et al. (1990)					
Al	101	39	145	25	145
Si	101	82	122	59	218
Calabrese et al. (1997)					
BTM[b]	256	7	74	20	73
Al	64	3	96	−3	67
Si	64	−16	57	−18	38
Studies of adults					
Calabrese et al. (1990)					
Al	6	77	65	57	
Si	6	5	55	1	
Stanek et al. (1997)					
BTM	10	6	165	−11	201
Al	10	12	31	5	
Si	10	−20	37	−24	
Davis and Mirick (2006)					
Mothers (Al)	19	92	218	0	
Mothers (Si)	19	23	37	5	
Fathers (Al)	19	68	130	23	
Fathers (Si)	19	26	49	0.2	
This study					
Al, Ce, La, Si	159	75	120	50	211
Al	43	37	52	31	110
Si	30	49	74	40	145

[a] LTM is the lowest soil ingestion rate of estimates generated from Al, Ti or Acid Insoluble Residue tracers and corrected for ingestion of tracers consumed in food and medicine.

[b] BTM is the best tracer method where the soil ingestion estimate is based on the median of values calculated for the 4 tracers with the lowest food/soil ratio.
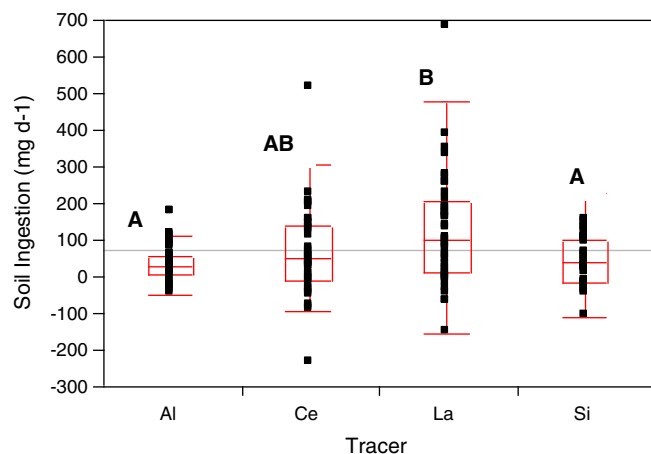

Fig. 2. Box plots of the frequency distributions of daily soil ingestion estimates for all study subjects for the 4 most reliable tracers. Significant differences in distribution means (Welch ANOVA, Tukey–Kramer HSD, p = 0.003) are denoted by differing labels.
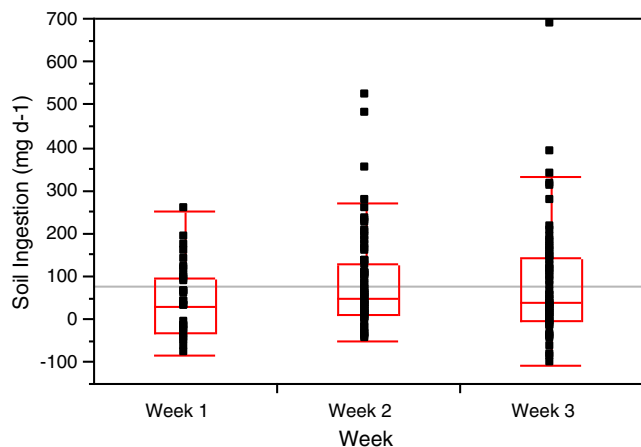
Fig. 3. Box plots of daily soil ingestion rate distributions calculated using the Al tracer for each study week of the Nemiah study showing median, 25% and 75% quantiles and outliers. The distributions are not significantly different (ANOVA F = 0.43, p = 0.65; Kruskall–Wallis test $\chi^2 = 0.45$, p = 0.80).

respectively, compared to values of 168%, 74%, 99% and 68%, respectively, for these tracers in the Stanek et al. (1997) study of adults.

As previously discussed, the 4 most reliable tracers used in this study are poorly absorbed in the gastrointestinal tract (assumption "c"). Specifically, gastrointestinal absorption factors (i.e., f_1 or the mass of tracer absorbed divided by the mass of tracer ingested) used in pharmacokinetic modeling of radionuclides in humans recommend f_1 values of 0.01, 0.005, 0.005 and 0.01, for Al, Ce, La and Si, respectively (ICRP, 1996). Moreover, Al and Si, originating from Plagioclase minerals, (i.e., $NaAlSi_3O_8$ and $CaAl_2Si_2O_8$) in the bedrock found in the study area are insoluble or only sparingly soluble in water (CRC, 1993).

The uncertainty related to source errors (i.e., assumption "d") was minimized by virtue of conducting the study under controlled conditions in a remote location, where the potential for problems due to unmeasured tracer sources (e.g., newspaper ink, urban dust and exhausts containing metals), and unrecorded food items (e.g., candies, snacks) were eliminated. The levels of the most reliable tracers used in the study were readily measurable in soils, and the variability in soils was generally observed to be low at each site (assumption "e"). The CV values were less than 5% for the 4 most reliable tracers at Henry's Crossing (Location C), where 2 of the 3 weeks of the study were completed, and at Farwell Canyon. The Al concentration in soil samples from the Nemiah Valley was observed to be an exception, with a CV value of 43%. However, this variability would only be expected to affect one subject for 1 week of the study. Soil ingestion estimate uncertainty was further reduced by calculating the soil ingestion rates using the concentration or depletion of tracers in the <63 μm soil fraction, thereby reducing the impact of tracer enrichment in the smaller particle size fractions that will be preferentially ingested.

Because of the potential for transit time misalignment of tracer inputs and tracer outputs, the ability to detect low levels of soil ingestion via the mass balance tracer method is encumbered by factors that increase the "signal to noise" ratio (Stanek et al., 2001). For example, sampling and analytical variability of food, fecal, and soil samples alone will result in a minimum detection level of 20 mg d^{-1}, according to estimates for ^{214}Pb using 225 subject days in a hypothetical mass balance tracer study (Doyle et al., 2010). Other factors that potentially contribute to the "noise" include variability of tracer levels in consumed foods, and/or variability in actual soil ingestion over the duration of the study within or between subjects. The negative effect of these factors on the precision of mass balance tracer soil ingestion estimates can be reduced by increasing the duration of the study, thereby
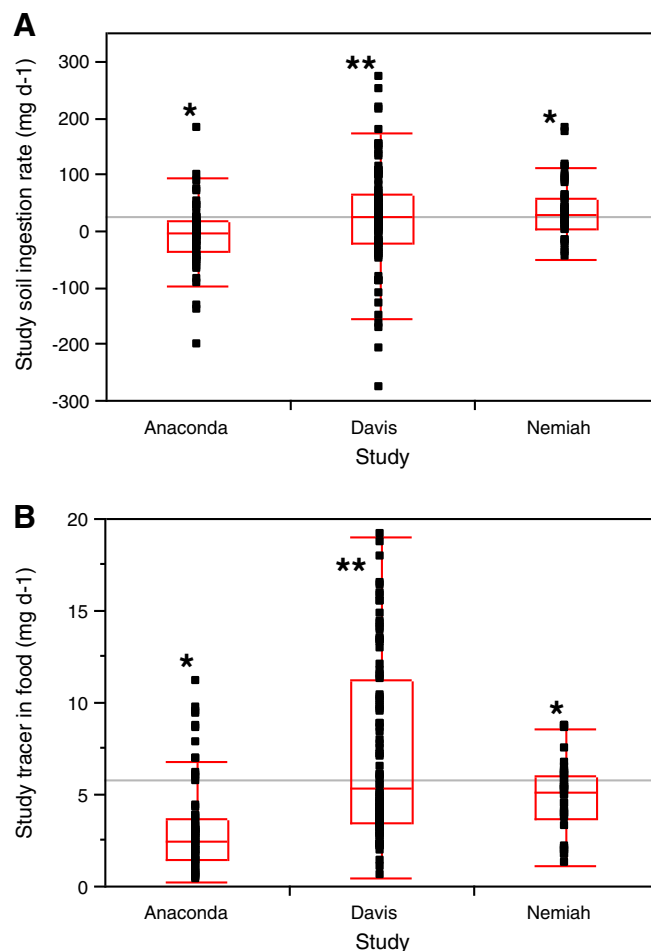

Fig. 4. A and B. Box plots of A) the frequency distributions of daily soil ingestion estimates, and B) frequency distributions of daily Al tracer ingestion rates in food, using the Al tracer results from the Anaconda study (Calabrese et al., 1997), the Davis study (Davis et al., 1990) and this study (Nemiah study). The box plots show the median, 25% and 75% quantiles, and outliers (i.e., 1.5 times the 25–75% inter-quartile value). The *'s correspond to variances that are unequal (Levene test F = 2.95, p = 0.054).

diluting the impact of transit time misalignment, and/or increasing the number of study subjects. However, implementing such changes would substantially increase the required cost and workload; moreover, a substantial increase in subjects is simply not possible for very small populations. However, in the current study variability in the levels of tracers in foods were reduced by providing a uniform diet over the duration of the study for all subjects, and by ensuring that all subjects lived under the same environmental conditions and participated in similar activities. The distributions of soil ingestion rate estimates and the distributions of daily tracer intake for Al observed in the Calabrese et al. (1997), Davis et al. (1990) and this study (i.e., Anaconda, Davis and Nemiah studies, respectively) of children are shown in Fig. 4A and B. The variance of soil ingestion rate estimates based on the Al tracer was observed to be unequal between the 3 studies (Levene test F = 2.95, p = 0.054). This difference in variance is reflected in the standard deviations on the mean soil ingestion rates reported for the Davis, Anaconda and Nemiah studies of 145 mg d^{-1}, 96 mg d^{-1} and 52 mg d^{-1}, respectively. Similarly, the variance in the daily Al intake in food by subjects in the 3 studies the variance were unequal (Levene test F = 30.1, p = 0.0001). Thus, as implied by using tracers with low F/S ratios,

lower variance in tracer intake in food results in lower variance (i.e., higher precision) in the estimated soil ingestion rate distribution.

This study, as with previous soil ingestion studies, has exhibited a high degree of variability between soil ingestion estimates calculated using different tracer elements. Meta-analysis of previous soil ingestion studies concluded that Al and Si are the most reliable mass balance tracers (Stanek et al., 2012). Similarly, the 2 most reliable tracers in this study were Al and Si because based on having the lowest F/S ratios of the tracers used. Further, both Al and Si found in local Plagioclase derived soils are sparingly soluble and thus poorly absorbed in the gastrointestinal tract, with f_1 values of 0.01. Moreover, the variability in the levels of Al and Si consumed in food during this study was observed to be relatively low, with CV values of 41% and 48%, respectively, thus reducing the "signal to noise" ratio for these tracers. This low variability is reflected in the low variance of estimate distributions calculated using these tracers.

5. Conclusions

This study represents the first quantitative soil ingestion study of a Canadian population, and the first of a population following a traditional wilderness lifestyle. Furthermore, this study is one of only a few quantitative soil ingestion studies of adults. As such, it constitutes an important contribution to the available research on this poorly understood exposure pathway used for the HHRAs of contaminated sites.

The mean and median soil ingestion rates measured in this study (i.e., 75 mg d^{-1} and 50 mg d^{-1} for the 4 most reliable tracers) are comparable or higher than the rates currently recommended by regulatory agencies for adults, based on reasonable central estimates of soil ingestion, which range from 20 mg d^{-1} to 100 mg d^{-1}. Moreover, the 90th percentile of the distribution of measured soil ingestion rates is in the order of 100 to 200 mg d^{-1}, and thus, much higher than those recommended for HHRAs of contaminated sites. Thus, the results from this study support the hypothesis that members of a community living in a rural and wilderness area who practice a traditional subsistence lifestyle experience higher soil ingestion rates than adults living in suburban/urban environments. This difference is substantial, but nevertheless, is less than the 400 mg d^{-1} scenario used to underpin qualitative exposure assessments for communities in the North-western United States that follow a similar lifestyle and estimates for construction workers or military personnel. However, the participants' activities during this study included mostly medium contact activities such as outdoor camping, hunting, and fishing, and further studies of adults would be required to determine if high-contact activities such as root digging and cultivating appreciably increase soil ingestion rates. Moreover, assuming that children are more susceptible to inadvertent soil ingestion, further studies of children living under rural and wilderness conditions, and involved traditional activities, are required to determine if soil ingestion rates currently recommended for HHRAs of contaminated sites are adequately protective for this receptor.

The study design, which included a group of subjects living under the same environmental conditions and consuming the same array of food items, reduced the variability of tracer intake from food and the mass of fecal output. As a result, the variance values associated with the soil ingestion rates estimated in this study were substantially lower than those of previous mass balance tracer studies (i.e., improved precision). This variability could be further reduced by eliminating processed foods that contain commonly used mass balance tracers, such as Al, from the subject's diet during the study. Al and Si were the most reliable tracers used in this study, based on their low F/S ratios, and the low variance of calculated soil ingestion rate estimates.

References

Binder S, Sokal D, Maughan D. Estimating soil ingestion: the use of tracer elements in estimating the amount of soil ingested by young children. Arch Environ Health 1986;41:341–5.

Calabrese EJ, Stanek EJ. An improved method for estimating soil ingestion in children and adults. J Environ Sci Health 1993;A28:363–71.

Calabrese EJ, Barnes R, Stanek EJ, Pastides H, Gilbert CE, Veneman P. How much soil do young children ingest: an epidemiologic study. Regul Toxicol Pharmacol 1989;10: 123–37.

Calabrese EJ, Stanek EJ, Gilbert CE, Barnes RM. Preliminary adult soil ingestion estimates: results of a pilot study. Regul Toxicol Pharmacol 1990;12:88–95.

Calabrese EJ, Stanek EJ, Pekow P, Barnes RM. Soil ingestion estimates for children residing on a superfund site. Ecotoxicol Environ Saf 1997;36:258–68.

Clausing P, Brunekreef B, van Wijnen JH. A method for estimating soil ingestion by children. Int Arch Occup Environ Health 1987;59:73–82.

CRC. In: Lide DR, editor. Handbook of Chemistry and Physics. 74th ed. Boca Raton, Fla: Chemical Rubber Company publishing; 1993.

Davis S, Mirick DK. Soil ingestion in children and adults in the same family. J Expo Sci Environ Epidemiol 2006;16:63–75.

Davis S, Waller P, Buschbom R, Ballou J, White P. Quantitative estimates of soil ingestion in normal children between the ages of 2 and 7 years: population-based estimates using aluminum, silicon, and titanium as soil trace elements. Arch Environ Health 1990;45:112–22.

deSilva PE. How much soil do children ingest—a new approach. Appl Occup Environ Hyg 1994;9:40–3.

Doyle JR, Blais JM, White PA. Mass balance soil ingestion estimating methods and their application to inhabitants of rural and wilderness areas: a critical review. Sci Total Environ 2010;408:2181–8.

Environment Canada. Canadian climate normals. Tatlayoko Lake RCS, British Columbia; National Climate Data and Information Archive; 1971–2000. Accessed May 2011; URL:www.climate.weatheroffice.gc.ca.

EPA. Exposure factors handbook. United States Environmental Protection Agency manual; 1997. Accessed February 2009. URLhttp://www.epa.gov/ncea/efh/pdfs/efh-front-gloss.pdf.

EPA. Supplemental guidance for developing soil screening levels for superfund sites. Environmental Protection Agency Report (OSWER 9355.4-24); 2002. URL:http://www.epa.gov/superfund/health/conmedia/soil/pdfs/ssg_main.pdf.

EPA. Exposure factors handbook. United States Environmental Protection Agency Manual Update (EPA/600/R-09/052a) Draft Issued for Public Review; 2009. Accessed February 2010. URL:http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=209866.

GSC. Chilko Lake and vicinity: coast and Lillooet districts. Geological Survey of Canada Department of Mines Bureau of Economic Geology document no. 2063; 1935.

Harper BL, Flett B, Harris S, Abeyta C, Kirschner F. The Spokane Tribe's multipathway subsistence exposure scenario and screening level reasonable maximum exposure. Risk Anal 2002;22:513–26.

Harper BL, Harding AK, Waterhous T, Harris SG. Traditional tribal subsistence exposure scenario and risk assessment guidance manual; appendix 2: soil ingestion rate. United States Environmental Protection Agency Report of Grant Number EPA-STAR-J1-R831046; 2005.

ICRP. Age-dependent doses to the members of the public from intake of radionuclides part 5; compilation of ingestion and inhalation coefficients. Annals of the ICRP, 72. ICRP Publication; 1996. p. 1-91.. 26.

Leet DL, Judson S. Physical geology. 4th ed. New Jersey: Prentice-Hall; 1971.

Madsen JL, Jensen M. Gastrointestinal transit of technetium-99m-labeled cellulose fiber and indium-I I l-labeled plastic particles. J Nucl Med 1989;30:402–6.

Plumlee GS, Ziegler TL. The medical geochemistry of dusts, soils, and other earth materials. In: Lollar BS, editor. Treatise on Geochemistry, vol 9. Amsterdam: Elsevier Publications; 2006.

Schiarizza P, Melville DM, Riddell J, Jennings BK. Geology and mineral occurrences of the Tatlayoko Lake map are (92 N/8, 9 and 10). British Columbia Geological Survey Branch Geological Fieldwork paper 1995-I; 1995.

Simon SL. Soil ingestion by humans: a review of history, data, etiology with application to risk assessment of radioactively contaminated soil. Health Phys 1998;74: 647–72.

Stanek EJ, Calabrese EJ. A guide to interpreting soil ingestion studies I: development of a model to estimate the soil ingestion detection level of soil ingestion studies. Regul Toxicol Pharmacol 1991;13:263–77.

Stanek EJ, Calabrese EJ, Barnes R, Pekow P. Soil ingestion in adults: results of a second pilot study. Ecotoxicol Environ Saf 1997;36:249–57.

Stanek EJ, Calabrese EJ, Zorn M. Biasing factors for simple soil ingestion estimates in mass balance studies of soil ingestion. Hum Ecol Risk Assess 2001;7:329–55.

Stanek EJ, Calabrese EJ, Xu B. Meta-Analysis of Mass-Balance Studies of Soil Ingestion in Children. Risk Anal 2012;32:433–47.

Supreme Court of British Columbia. Tsilhqot'in Nation v. British Columbia. 2007-BCSC-1700; 2007.

UMass. Soil ingestion in children-meta analysis website. Amherst: University of Massachusetts; 2005. Accessed March 2011; URL:http://www.umass.edu/metasoil/.

Umhoefer PJ, Schiarizza P, Robinson M. Relay mountain group, Tyaughton–Methow basin, southwest British Columbia: a major Middle Jurassic to Early Cretaceous terrane overlap assemblage. Can J Earth Sci 2002;39:1143–67.

USAPHC. Methodology for developing chemical exposure guidelines for deployed military personnel. U.S. Army Public Health Command (USAPHC) Technical Guide Reference Document 230, June 2010 Revision, developed in support document for Technical Guide 230; 2010.

Van Holderbeke M, Cornelis C, Bierkens J, Torfs R. Review of the Soil Ingeston Pathway in Human Exposure Assessment. Study in support of the BenNeKempen project, Flemish Institute for Technological Research (VITO); 2007.

van Wijnen JH, Clausing P, Brunekreef B. Estimated soil ingestion by children. Environ Res 1990;51:147–62.

Wilson Consulting, Meridian Environmental. Critical review of soil ingestion rates for use in contaminated site human health risk assessments in Canada. Health Canada report prepared for the Safe Environments Programme, Health Canada; 2006.

Xeni Gwet'in. Xeni Gwet'in the people of Nemiah: about Nemiah. Xeni Gwet'in First Nation Government; 2011. Accessed June 2011; URL:http://www.xenigwetin.com/.